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Modulation Broadening of Unsaturated Lorentzian Lines*

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Closed, analytic expressions are obtained for the harmonic amplitudes which arise in the modulation of unsaturated Lorentzian absorption lines. Exact formulas relating characteristics of the observed signals (amplitude, width, slope ratios, etc.) to the true half-width for arbitrary modulation amplitude are derived. The results of greatest experimental interest are graphed.

IN the interpretation of magnetic resonance data obtained with phase-detection techniques, it is necessary to allow for the effect of modulation broadening on the absorption lines. Several authors[1] have discussed this problem and supplied useful corrections to observed quantities. These formulas, however, have been either approximate or based on series expansions whose convergence is slow in some regions of experimental interest. The series in fact do not converge at all over certain ranges of modulation amplitude. In connection with an experimental program requiring accurate corrections for large amplitude modulation, the effect of modulation broadening on unsaturated Lorentzian lines was calculated for arbitrary modulation amplitude. The results are in closed form and are exact for the case of very slow sweep through the line.

Let $H_a(t)$ be the homogeneous applied magnetic field whose time dependence involves only the slow linear sweep across an absorption line. Let H_0 be the field at which resonance occurs, $H_{\frac{1}{2}}$ the half-width (distance between half-intensity points) of the true line, and H_ω the amplitude of the sinusoidal modulation with circular frequency ω. The normalized unsaturated Lorentzian absorption line may be written

$$g(H)=\pi^{-1}\frac{\frac{1}{2}H_{\frac{1}{2}}}{(\frac{1}{2}H_{\frac{1}{2}})^2+(H-H_0)^2}, \tag{1}$$

and under modulation a signal will be generated which is proportional to

$$g[H(t)]=\pi^{-1}\frac{\frac{1}{2}H_{\frac{1}{2}}}{(\frac{1}{2}H_{\frac{1}{2}})^2+[H_a(t)+H_\omega\cos\omega t-H_0]^2}. \tag{2}$$

The sweep rate is assumed to be very small so that $H_a(t)$ remains essentially constant over a time interval $2\pi/\omega$. Writing $H_a-H_0=H_\delta$ and Fourier analyzing $g(t)$,

$$\begin{aligned} g(t)&=\pi^{-1}\frac{\frac{1}{2}H_{\frac{1}{2}}}{(\frac{1}{2}H_{\frac{1}{2}})^2+(H_\delta+H_\omega\cos\omega t)^2} \\ &=\frac{H_{\frac{1}{2}}}{2\pi}\sum_{n=0}^{\infty}a_n(H_{\frac{1}{2}},H_\omega,H_\delta)\cos n\omega t, \end{aligned} \tag{3}$$

where the integrals for the Fourier amplitudes

$$\begin{aligned} &a_n(H_{\frac{1}{2}},H_\omega,H_\delta) \\ &\quad=(\omega/\pi)\int_{-\pi/\omega}^{\pi/\omega}\frac{\cos n\omega t}{(\frac{1}{2}H_{\frac{1}{2}})^2+(H_\delta+H_\omega\cos\omega t)^2}dt \end{aligned} \tag{4}$$

may be performed by a standard technique of contour integration.[2] Using phase detection of the fundamental, the recorded signal will be proportional to the Fourier coefficient a_1. Since the integration may be performed at once for all n, an expression for the amplitude of any harmonic will be displayed. This is then specialized to the case of primary interest here, and the properties of a_1 further investigated.

Define dimensionless parameters α and β where

$$\alpha=(H_\delta/H_\omega)\quad -\infty<\alpha<\infty\qquad \beta=(\tfrac{1}{2}H_{\frac{1}{2}}/H_\omega)\quad 0<\beta<\infty \tag{5}$$

and the auxiliary variables γ, u, r, and ϕ, where

$$\begin{aligned} &\gamma=1+\beta^2+\alpha^2, \\ &u=\gamma+[\gamma^2-4\alpha^2]^{\frac{1}{2}} &&2<u<\infty. \\ &r=[u-1-u^{\frac{1}{2}}(u-2)^{\frac{1}{2}}]^{\frac{1}{2}} &&0<r<1, \\ &\phi=\arc\cos\{-\sqrt{2}\alpha/u^{\frac{1}{2}}\} &&0<\phi<\pi. \end{aligned} \tag{6}$$

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[1] M. M. Perlman and M. Bloom, Phys. Rev. **88**, 1290 (1952); E. R. Andrew, *ibid.* **91**, 425 (1953); R. Beringer and J. G. Castle, *ibid.* **81**, 82 (1951); O. E. Myers and E. J. Putzer, J. Appl. Phys. **30**, 1987 (1959).

[2] P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill Book Company, Inc., New York, 1953), Vol. 1, p. 408.

The result of the integration for any n is

$$a_n(H_{\frac{1}{2}}, H_\omega, H_\delta)=\left(\frac{2}{H_\omega}\right)^2\left\{\frac{[r^{n-1}+r^{-(n-1)}]\sin(n+1)\phi-[r^{n+1}+r^{-(n+1)}]\sin(n-1)\phi}{[r^{-1}-r][r^2+r^{-2}+2\cos 2\phi]\sin\phi}+I_n\right\}, \tag{7}$$

where

$$I_0=I_1=0,$$

$$I_2=1,$$

$$I_n=\left\{(d/dx)^{n-2}\left[\frac{1}{(x-z)(x-z^*)[x-(1/z)][x-(1/z^*)]}\right]\right\}_{x\to 0}, \tag{8}$$

$$z=r\exp(i\phi).$$

For the cases $n=0, 1, 2$ the general expression reduces to

$$a_0=\left(\frac{2}{H_\omega}\right)^2\frac{u^{\frac{1}{2}}}{2(u-2)^{\frac{1}{2}}(u-\gamma)}, \tag{9}$$

$$a_1=\pm\left(\frac{2}{H_\omega}\right)^2\frac{(2\gamma-u)^{\frac{1}{2}}}{2(u-2)^{\frac{1}{2}}(u-\gamma)}, \tag{10}$$

$$a_2=\left(\frac{2}{H_\omega}\right)^2\left[1+\frac{u^{\frac{1}{2}}(1+2\gamma-2u)}{2(u-2)^{\frac{1}{2}}(u-\gamma)}\right]. \tag{11}$$

The detected signal, $a_1[H_{\frac{1}{2}}, H_\omega, H_\delta(t)]$, is obtained by restoring the linear time variation of H_a, or equivalently, H_δ. The pertinent properties of the resulting curve, which is similar in shape to the derivative of the Lorentzian curve, may be obtained by taking the derivative

$$(da_1/dH_\delta)=-(2/H_\omega^3)\frac{u^{\frac{1}{2}}(u^2-u-2\gamma u+3\gamma)}{(u-2)^{\frac{1}{2}}(u-\gamma)^3}. \tag{12}$$

Setting the factor $(u^2-u-2\gamma u+3\gamma)$ to zero generates relationships giving the location and amplitude of the two anti-symmetric peaks of a_1 for any modulation amplitude. Letting the symbol for any quantity with a suffix p attached denote that quantity evaluated at the peaks, these relations are

$$(H_\delta)_p=\alpha_p H_\omega=(\alpha_p/2\beta)H_{\frac{1}{2}}, \tag{13}$$

and

$$(a_1)_p=\pm\tfrac{3}{2}(2/H_{\frac{1}{2}})^2\left[\frac{(u_p-2)}{u_p(2u_p-3)}\right]^{\frac{1}{2}} \tag{14}$$

$$\alpha_p=\pm[1+\tfrac{5}{3}\beta^2-\tfrac{4}{3}\beta(\beta^2+\tfrac{3}{4})^{\frac{1}{2}}]^{\frac{1}{2}} \tag{15}$$

$$u_p=2+\tfrac{4}{3}\beta^2+\tfrac{4}{3}\beta(\beta^2+\tfrac{3}{4})^{\frac{1}{2}}. \tag{16}$$

Additional expressions, which often facilitate manipulations, are

$$\gamma_p=\frac{u_p(u_p-1)}{2u_p-3}\qquad 4\alpha_p^2=\frac{u_p^2}{2u_p-3}\qquad 4\beta^2=\frac{3(u_p-2)^2}{2u_p-3}. \tag{17}$$

Figure 1 illustrates the dependence of the location and amplitude of the peaks on the modulation amplitude.

Setting the derivative $(d/dH_\omega)[(a_1)_p]$ to zero, it further results that the maximum possible height of the peaks of a_1 occurs when $\beta=\frac{1}{2}$. On letting the subscript m indicate fulfillment of this condition, the following values are obtained:

$$\alpha_{pm}=\pm\tfrac{1}{2}\sqrt{3}\qquad \gamma_{pm}=2\qquad u_{pm}=3$$

$$(H_\omega)_m=H_{\frac{1}{2}}\qquad (H_\delta)_{pm}=\pm\sqrt{3}/2(H_{\frac{1}{2}})$$

$$(a_1)_{pm}=\pm 2(1/H_{\frac{1}{2}})^2. \tag{18}$$

These results are contained implicitly in Fig. 1. Quantities corresponding to $(H_\delta)_p$ and $(a_1)_p$ for an unbroadened line would be the location $(H_\delta)_{pL}$ and height $(a_1)_{pL}$, of the peaks in the first derivative of a pure Lorentzian curve. Calculation shows that

$$(H_\delta)_{pm}=3(H_\delta)_{pL}\qquad (a_1)_{pm}=(4\pi/3\sqrt{3})(a_1)_{pL}. \tag{19}$$

Another experimentally useful characteristic is the ratio of maximum slopes of a_1. Equating the second derivative

$$d^2a_1/dH_\delta^2=\pm(6/H_\omega^4)\times\frac{(2\gamma-u)^{\frac{1}{2}}[(\gamma-1)u^3-(\gamma-1)(1+2\gamma)u^2+4\gamma(\gamma-1)u-\gamma^2]}{(u-2)^{\frac{1}{2}}(u-\gamma)^5} \tag{20}$$

to zero gives the possibilities

$$u=2\gamma, \tag{21}$$

or

$$(\gamma-1)u^3-(\gamma-1)(1+2\gamma)u^2+4\gamma(\gamma-1)u-\gamma^2=0. \tag{22}$$

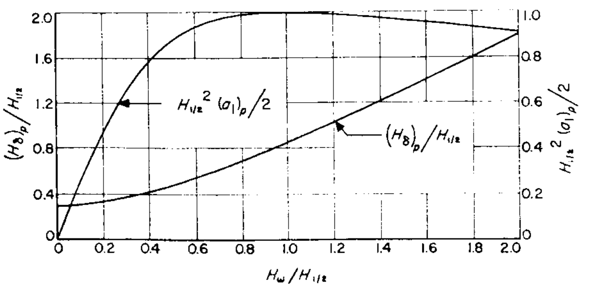

FIG. 1. Dependence of the location $(H_\delta)_p$ and height $(a_1)_p$ of the observed peaks on modulation amplitude.

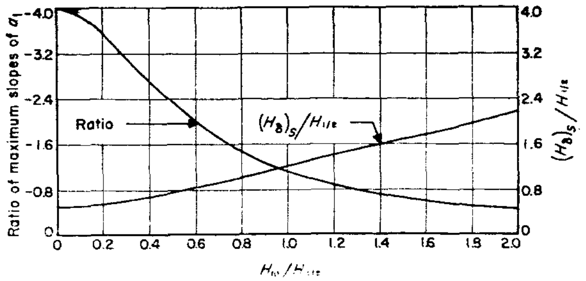

FIG. 2. Dependence of the location of outer maximum slope $(H_\delta)_s$ and the ratio of maximum slopes on modulation amplitude.

The first of these equations implies $H_\delta=0$ from the definition of u. Evaluating Eq. (12), the inner maximum slope of a_1 occurring at $H_\delta=0$ is in general,

$$da_1/dH_\delta\,|_{H_\delta=0}=-2(2/H_{\frac{1}{2}})^3[\beta^2/(1+\beta^2)^{\frac{3}{2}}]. \quad (23)$$

The outer maximum slope is given by one of the roots of the equation

$$(\gamma_s-1)u_s^3-(\gamma_s-1)(1+2\gamma_s)u_s^2+4\gamma_s(\gamma_s-1)u_s-\gamma_s^2=0, \quad (24)$$

where the subscript s indicates the value of a quantity at the place of outer maximum slope. It is quite cumbersome to extract this root straightforwardly, except for the case where $\beta=\frac{1}{2}$ which is discussed below. For the general case, an indirect approach which proceeds as follows seems most feasible. Treating Eq. (24) as a quadratic for γ_s, it is solved to obtain

$$\gamma_s=\frac{u_s}{2(2u_s^2-4u_s+1)}\times\{u_s^2+u_s-4+(u_s-2)(u_s^2-2u_s+5)^{\frac{1}{2}}\}. \quad (25)$$

After selecting a value for u_s, γ_s is fixed by this equation. Then α_s and β are evaluated with the equations

$$\alpha^2=\tfrac{1}{4}u(2\gamma-u) \quad (26)$$

$$\beta^2=\gamma-1-\alpha^2, \quad (27)$$

which follow immediately from the definitions of γ and u. The slope ratio can then be found using Eqs. (12) and (23). Figure 2, showing how the location of outer maximum slope and the slope ratio depend on modulation amplitude, was constructed using this procedure.

Returning to the case when a_1 has maximum amplitude, i.e., $\beta=\frac{1}{2}$, it can be shown that Eq. (24) reduces to

$$u_{sm}^3-5u_{sm}^2+3u_{sm}+5=0, \quad (28)$$

where the subscript m means simply $\beta=\frac{1}{2}$ as before. The only valid root of this equation is expressible as

$$u_{sm}=\tfrac{1}{3}[5+8\cos\{\pi/6+\tfrac{1}{3}\sin^{-1}(5/32)\}]\cong 3.9032, \quad (29)$$

and the other quantities needed are given in terms of u_{sm} by

$$\gamma_{sm}=\frac{u_{sm}^2-5}{2(u_{sm}-2)}\cong 2.6888,$$

$$\alpha_{sm}=\pm\left[\frac{u_{sm}(u_{sm}-\frac{5}{2})}{2(u_{sm}-2)}\right]^{\frac{1}{2}}\cong\pm 1.1995$$

$$(H_\delta)_{sm}=\alpha_{sm}H_{\frac{1}{2}} \quad (30)$$

$$\left(\frac{da_1}{dH_\delta}\right)_{sm}=\left(\frac{2}{H_{\frac{1}{2}}}\right)^3\frac{u_{sm}^{\frac{1}{2}}(u_{sm}-2)^{\frac{1}{2}}(3u_{sm}-5)(u_{sm}-3)}{(u_{sm}^2-4u_{sm}+5)^3}\cong 0.31833(2/H_{\frac{1}{2}})^3.$$

Evaluating Eq. (23) for $\beta=\frac{1}{2}$,

$$(da_1/dH_\delta)_{H_\delta=0}=-(4/5(5)^{\frac{1}{2}}](2/H_{\frac{1}{2}})^3\cong -0.35777(2/H_{\frac{1}{2}})^3, \quad (31)$$

and so the slope ratio for $\beta=\frac{1}{2}$ is -1.1239. In comparison, the slope ratio is -4 for the derivative of the Lorentz curve.

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